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### A composite resin composition

Hitherto, there have been proposed and commercialized many kinds of composite composition of thermoplastic resins. As the additive used therein, inorganic fillers such as calcium carbonate, talc, mica and the like are well known. Moreover, there are also known organic materials such as wood flour, chaff and the like; fibrous materials such as asbestos and glass fiber; as well as graphite, cokes, blown asphalt, activated carbon, magnesium hydroxide, aluminum hydroxide and the like. However, these additives have high specific gravities, and their ability to improve physical property of composition is not so sufficient. The present inventor found that, by incorporating a flax or flax-like fibers into a thermoplastic resin, physical properties of the resin, particularly its dimensional stability, heat distortion temperature, stiffness, etc., can be improved surprisingly and further its coatability can be improved and its tapping strength can be made sufficient. After additional studies, this invention was accomplished.

The term "flax" referred to in this invention means flax, ramie, hemp, jute, Manila hemp, sisal and the like, and the term "flax-like fibre" referred to in this invention means Indian mallow, white flax, kenaf, raffia fiber, New Zealand flax, coir fiber, hemp-palm fiber and the like.

(1) Coating process, i.e. a process which comprises coating the surface of filler with a substance such as surfactant, (such as waxy) fatty acid and its salt, or the like, or with a substance having compatibility with the resin and excellent in adhesion to the filler such as rubber type or vinyl type of low molecular weight materials or the like;

(3) Surface treatment with a polymerizable monomer, functional oligomer, functional polymer or the like, i.e., a process which comprises treating the surface of a filler with acrylic acid, maleic acid, maleic anhydride, liquid polybutadiene or other rubber type oligomer having terminal carboxyl group, and a polymer modified with maleic anhydride, carboxyl group-containing polyolefin or the like, and a process which comprises adding an organic peroxide on heating the filler in the course of the above-mentioned surface treatment.

A combined use of the filler of this invention with known filler or additive with consideration of cost and properties of the composite is naturally possible, and such a combined use is also included in the technical scope of this invention.

In the following examples, the case of Jute will be mentioned as the most typical example.

EXAMPLE 1

Jute fibers were cut into a fiber length of 5 mm or less and an average length of about 3 mm. The cut fibers of which surface had been treated with the treating agent mentioned later in Table 3 or without any treatment were added to polypropylene resin. The resulting mixture was then roughly treated on hot rolls, roughly ground, and then formed into test pieces by means of an injection molding

machin . Several properties f the test pieces wer measured acc rding to the ASTM (Americal Society  
f r Testing and Materials) methods indicated below:

	Tensile Strength	—	ASTM D—638	
	Flexural Strength	—	ASTM D—790	
5.	Flexural Modulus	—	ASTM D—648	5
	Hardness	—	ASTM D—785	
	Izod impact strength	—	ASTM D—256	

TABLE 1

Polypropylene (MA-3) (parts)		100	90	80	70	60	50	40
Jute, (untreated) (parts)		0	10	20	30	40	50	60
Hardness, R scale		101	102	104	106	110	110	103
Izod impact strength (kg.cm/cm <sup>2</sup> )		4.5	4.5	4.7	4.6	4.6	4.6	4.0
Tensile strength (kg/cm <sup>2</sup> )		368	380	385	408	524	388	370
Dimension	Width (mm)	48.8	48.8	49.0	49.2	49.2	49.2	49.2
	Length (mm)	88.4	88.5	88.7	89.0	89.3	89.3	89.3

(Notes) MA-3: Trademark for a homo-grade polypropylene, manufactured by Mitsubishi Petrochemical Company Limited.

Heat distortion temperatures: MA-3 itself has a heat distortion temperature of 120°C, and the heat distortion temperature becomes 140°C when 20 — 40 parts of jute is added to 80 — 60 parts of MA-3 to give a total (jute + MA-3) of 100 parts.

TABLE 2

Polypropylene (B-200) (parts)	100	90	80	70	60	50	40
Jute, untreated (parts)	0	10	20	30	40	50	60
Hardness, R scale	89	88	91	91	97	97	97
Izod impact strength (kg.cm/cm <sup>2</sup> )	10	10	10	9	9	7	6
Tensile strength (kg/cm <sup>2</sup> )	355	355	360	401	430	430	342
Dimension	Width (mm)	48.8	48.9	49.0	49.1	49.1	49.1
	Length (mm)	88.3	88.4	88.9	89.1	89.2	89.2

(Notes) B-200: Trademark for a copolymer grade polypropylene, manufactured by Mitsui Toatsu Chemicals, Inc.

Heat Distortion temperatures: B-200 itself has a heat distortion temperature of 115°C, and the heat distortion temperature becomes 135°C when 20 - 40 parts of jute is added to 80 - 60 parts of B-200 to give a total (jute + B-200) of 100 parts.

TABLE 3

Polypropylene (B-200) (parts)	100	60	60	60	60	60
Surface-treating agent	—	Zinc stearate	Nipol LX 111	EPR EP-57P	A 1100 Silane	TTS Titanate
Jute, treated (parts)	0	40	40	40	40	40
Hardness, R scale	87	87	88	86	98	95
Izod impact strength (kg.cm/cm <sup>2</sup> )	10	7	9	9	7	6
Tensile strength (kg/cm <sup>2</sup> )	355	333	380	376	415	403
Dimension	48.8	49.2	49.0	48.9	49.1	49.0
	88.3	89.2	89.0	88.8	89.0	89.0

(Notes) Nipol LX 111: Trademark for a polybutadiene latex, manufactured by Nippon Zeon Corp.

EPR EP-57P: Trademark for a ethylene propylene rubber, manufactured by Mitsubishi Petrochemical & Co., Ltd.

A1100 (silane): Trademark for a silane coupling agent, manufactured by Nippon Unicar Co.

TTS (titanate): Trademark for a titanate coupling agent, supplied by Ajinomoto Co., Inc.

In the experiments of Table 3, the surface of the jute fibers was treated with the treating agent mentioned in Table 3 and then 40 parts of the treated jute fibers were added to 60 parts of polypropylene and mixed.

Details of the treating agents used in the experiments of Table 3 and their amounts (per cent by weight of jute) were as shown in the following Table 4.

TABLE 4

Surface-treating agent	Amount used in the treatment	Note
Zinc stearate (manufactured by Sakai Chemical Industry Co., Ltd.)	5%	The fibers were treated with 10% solution of the agent in toluene and then dried.
Nipol LX 111	5%	After the treatment, the fibers were dried.
EP-57P	5%	The fibers were treated with 10% solution of EPR in toluene and then dried.
A 1100 (Silane)	2%	The fibers were treated with aqueous solution of A1100 (diluted to 100 times) and then dried.
TTS (Titanate)	2%	The same as above.

In the experiments of Tables 1 and 2, the untreated fibers were added to polypropylene resin as shown in these tables, even in these experiments, the surface hardness and stiffness of polypropylene resin were markedly improved, and the poor dimensional stability-mold shrinkage- of the olefin resins was also improved greatly. At the same time, heat distortion temperature of the composite also rose by about 20°C (when 20—40 parts of the fibers were added).

Next, treating agent A or B (20 parts each) mentioned below was added to 100 parts of jute fibers cut to size of 1.5 mm, and the resulting mixture was thoroughly treated on hot rolls, cooled and roughly ground. Sixty parts of the ground mixture were mixed with 40 parts of polypropylene resin, and then molded pieces were prepared. Properties of the pieces were measured to obtain the results shown in Table 5 (Experiments A and B).

A: Admer QE 305, (trademark for an adhesive polyolefin resin containing small percentage of carboxyl group manufactured by Mitsui Petrochemicals Co., Ltd.)

B: Bondfast G (a trademark for an adhesive resin which is said to be modified with maleic anhydride for structural use manufactured by Sumitomo Chemical Co., Ltd.)

Next, a mixture consisting of 60 parts of polypropylene resin, 40 parts of cut jute having a size of 1.5 mm, 0.5 part of maleic anhydride and 0.1 part of dicumyl peroxide was extrusion molded (180—200°C), and then injection molded pieces were prepared. Properties of the pieces were measured to obtain the results shown in the following Table 5 (Experiment C).



TABLE 5

Polypropylene (MA-3)	100%	A	B	C	Polypropylene (B-200)	100%	A	B
Tensile strength (kg/cm <sup>2</sup> )	388	510	490	430	Tensile strength (kg/cm <sup>2</sup> )	355	480	430
Flexural strength (kg/cm <sup>2</sup> )	260	590	550	510	Flexural strength (kg/cm <sup>2</sup> )	250	490	500
Flexural modulus (kg/cm <sup>2</sup> )	11,600	48,000	42,000	3,900	Flexural modulus (kg/cm <sup>2</sup> )	8,900	43,000	38,000

Next, a mixture consisting of 100 parts of cut jute having a size of 1.5 mm and 20 parts of Admer QE 305 was treated on hot rolls and then roughly ground. The ground mixture was mixed with the ingredients shown in Table 6, extrusion molded, and then injection molded pieces were prepared. Properties of the pieces were measured to obtain the results shown in Table 6.

TABLE 6

Polypropylene (MA-3)	100	60	60	60
Calcium carbonate	0	10	0	0
Talc	0	0	10	0
Glass fiber	0	0	0	10
Surface-treated jute	0	30	30	30
Tensile strength (kg/cm <sup>2</sup> )	368	460	470	560
Flexural strength (kg/cm <sup>2</sup> )	260	430	510	620
Flexural modulus (kg/cm <sup>2</sup> )	11,600	36,000	51,000	57,000

When the surface of jute fibers was treated, there were obtained various products having excellent hardness, good dimensional stability, or high Izod impact strength.

## EXAMPLE 2

Jute fibers were added to styrene resin and the mixture was treated according to the procedure of Example 1. The results obtained were shown in the following tables. 10

TABLE 7

Styrene resin (parts)	100	95	90	80	70
Untreated jute (parts)	0	5	10	20	30
Hardness (R scale)	118	118	121	123	123
Izod impact strength (kg·cm/cm <sup>2</sup> )	4	4	4	4	4
Tensile strength (kg/cm <sup>2</sup> )	493	495	525	610	605

(Note) Styrene resin: Styrene 666 (manufactured by Asahi-Dow Limited).

TABLE 8

Styrene resin (parts)	100	100	100	100	100	100
Surface-treating agent	--	Zinc stearate	A 174 Silane	JSR 0592	9S Titanate	Acrylic AP-9240M
Treated jute (parts)	0	20	20	20	20	20
Hardness (R scale)	118	118	123	120	121	119
Charpy impact strength (kg cm/cm <sup>2</sup> )	4	4	4	4.8	4	4.6
Tensile strength (kg/cm <sup>2</sup> )	493	572	750	849	880	700

(Notes) JSR 0592: Trademark for a carboxyl-modified styrene-butadiene latex, manufactured by Japan Synthetic Rubber Co.

A174

Silane:

Trademark for a silane coupling agent, manufactured by Nippon Unicar Co.

9S

Titanate:

Trademark for a titanate coupling agent, supplied by Ajinomoto Co., Inc.

Acrylic

AP-9240M:

Trademark for an adhesive acrylic resin latex, supplied by Nippon Shokubai Kagaku Co., Ltd.

In the experiment of Table 8, the surface of jute fibers was treated with the agent of which name and amount (percent by weight of jute) are shown in Table 10, and then the treated jute fibers were added to styrene resin in the proportion shown in Table 8, and followed by mixing, extrusion and injection molding.

TABLE 9

Surface-treating agent	Amount used in the treatment	Note
Zinc-stearate (manufactured by Sakai Chemical Industry Co., Ltd.)	5%	The fibers were treated by the same procedure as in Example 1.
A 174 Silane	2%	The same as above.
JSR 0592	5%	The same as above.
9S Titanate	2%	The same as above.
Acrylic AP-9240M	5%	The fibers were treated with aqueous solution of AP-9240M (diluted to 100 times) and then dried.

**EXAMPLE 3**

By repeating the procedure of Example 1 according to the formulations shown in Tables 10, 11 and 12, jute fibers were added to SAN resin followed by mixing, extrusion and injection molding.

TABLE 10

SAN resin (parts)	100	80	80	80	80	80
Surface-treating agent	—	A 174 Silane	TTS Titanate	JSR 0592	Sumilite resin R-968	Epoxy R3360
Treated jute (parts)	0	20	20	20	20	20
Hardness (R scale)	121	123	122	121	122	121
Izod impact strength (kg-cm/cm <sup>2</sup> )	5.6	5	5	5	5.6	5
Tensile strength (kg/cm <sup>2</sup> )	770	833	819	739	903	785

(Note) SAN resin: SANREX C, Trademark for an acrylonitrile-styrene resin  
manufactured by Mitsubishi Monsanto Chemical Company.  
Sumilite Resin R-968: Trademark for a phenol resin (40% aqueous solution),  
manufactured by Sumitomo-Durez Co., Ltd.

Epoxy R 3360: Trademark for an epoxy resin, manufactured by Japan Reichhold  
Chemicals, Inc.

TABLE 11

Surface-treating agent	Amount used in the treatment	Note
A 174 Silare	2%	The same as in Table 9.
TTS Titanate	2%	The same as in Table 4.
JSR 0592	2%	The same as in Table 9.
Sumilite Resin R-968	5%	The fibers were treated with aqueous solution (diluted to 100 times) of the phenolic resin and then dried.
Epoxy R 3360	5%	The fibers were treated with aqueous solution (diluted to 100 times) of the epoxy compound and then dried.

TABLE 12

SAN resin (parts)	80	80	80	80	80	80
Amount of phenolic resin used in the treatment (%)	No treatment	2	5	10	15	20
Treated jute (parts)	20	20	20	20	20	20
Hardness (R scale)	111	111	112	112	111	111
Izod impact strength (kg. cm/cm <sup>2</sup> )	6	5	5.6	6	5	4
Tensile strength (kg/cm <sup>2</sup> )	847	860	903	915	900	796

Thus, 20 parts of jute fibers subjected to the respective treatment were added to 80 parts of the resin. As a result, tensile strength was improved to a great extent particularly.

5 Good results were obtained also in the case of other flax and flax-like fibers but jute.

Finally, additional inorganic materials, antistatic agent, lubricant, colorant, flame retardant, foaming agent and the like could be incorporated into the composite resin composition of this invention.

10 The "tapping strength" referred to earlier in this specification is the strength of a material around a hole being screwed with a tap: if the strength of the material is not high enough, then one or more cracks will be formed around the hole.

15 As indicated earlier, the term "flax or flax-like fibres" used in this specification means "bast fibers". The invention thus provides a composition comprising 10 parts by weight of a thermoplastic resin or blend of thermoplastic resins and 0.5 to 20 parts by weight of bast fibers having a fiber length of 10 mm or less, which fibers may or may not have undergone a surface treatment.

#### CLAIMS

1. A composite resin composition characterized by compounding 10 parts by weight of a thermoplastic resin or resin blend, 0.5—20 parts by weight of flax or flax-like fibers or a mixture thereof having a fiber length of 10 mm or less and with or without surface treatment.

20 2. A composite resin composition according to Claim 1, wherein said thermoplastic resin is a molding thermoplastic resin or resin blend, and the molding thermoplastic resin includes a homo polymer, copolymer and reactive modified resin selected from the group consisting of polyvinyl chloride (PVC), polypropylene (PP), polyethylene (PE), styrene resin, acrylonitrile-styrene resin (SAN) and acrylonitrile-butadiene-styrene resin (ABS).

3. A composite resin composition according to Claim 1 or claim 2, wherein said flax is at least one flax selected from the group consisting of flax, ramie, hemp, jute, Manila hemp, and sisal.

4. A composite resin composition according to any one of Claims 1 to 3 wherein said flax-like fiber is at least one flax-like fiber selected from the group consisting of Indian mall w, white flax, kenaf, raffia  
5 fiber, New Zealand flax, coir fiber and hemp-palm fiber. 5

5. A composition as claimed in claim 1 and described in any one of the Examples herein.

6. A shaped article whenever made from a composition as claimed in any one of claims 1 to 5.

7. Any new and novel feature described herein.